Supplemental Information for:

The Quantum Mechanics Derived Atomistic Mechanism Underlying the Acceleration of Catalytic CO oxidation on Pt(110) by Surface Acoustic Waves

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1. Computational details for DFT and AIMD simulations

1.1 Numerical set-up of the DFT calculations:

- Quantum Espresso (QE) code
- lattice parameter Pt-Pt = 2.82 Å = DFT relaxed (experiment 2.775 Å)
- PAW description of atomic core
- cut-offs typically 30/300 Ry
- smearing: 0.002-0.007 Ry
- NEB 5-9 images, k = 0.3-0.6, in 1-3 steps increasing accuracy
- spin-unrestricted
- unit cell = (3x2)
- # layers = 5, bottom 2 frozen
- dipole correction implemented[1]

Additionally, we predicted the work function for both the bare Pt(110) reconstructed surface and the unreconstructed surface at both low and high coverage of both O and CO:

coverage/system	unreconstructed	reconstructed
bare	5.36 eV	5.40 eV
O-covered	7.25 eV	6.14 eV
CO-covered	5.72 eV	6.35 eV

Experiments[2,3] determined that the work function of unreconstructed Pt(110) at full CO coverage is larger than the bare reconstructed surface by 0.15 eV, while the work function of reconstructed Pt(110) at full O coverage is larger than the bare reconstructed surface by 0.8 eV. These values are in reasonable comparison with our predictions: Δ(work function) = 0.32 eV for full CO coverage and Δ(work function) = 0.74 eV for full O coverage.

1.2 AIMD simulations

Our results show that several dynamic processes are enhanced in these simulations, such as:

- CO diffusion (also in the form of a Pt-CO unit as illustrated in Figure 3a)
- Pt-CO detachment (Figure 3b)
- COox reaction (Figure 3c)

For convenience of the reader we give as a Supplementary File a movie of an AIMD trajectory as an example of these simulations: in this movie a LH COox reaction occurs at an early stage (around 2.60 psec).

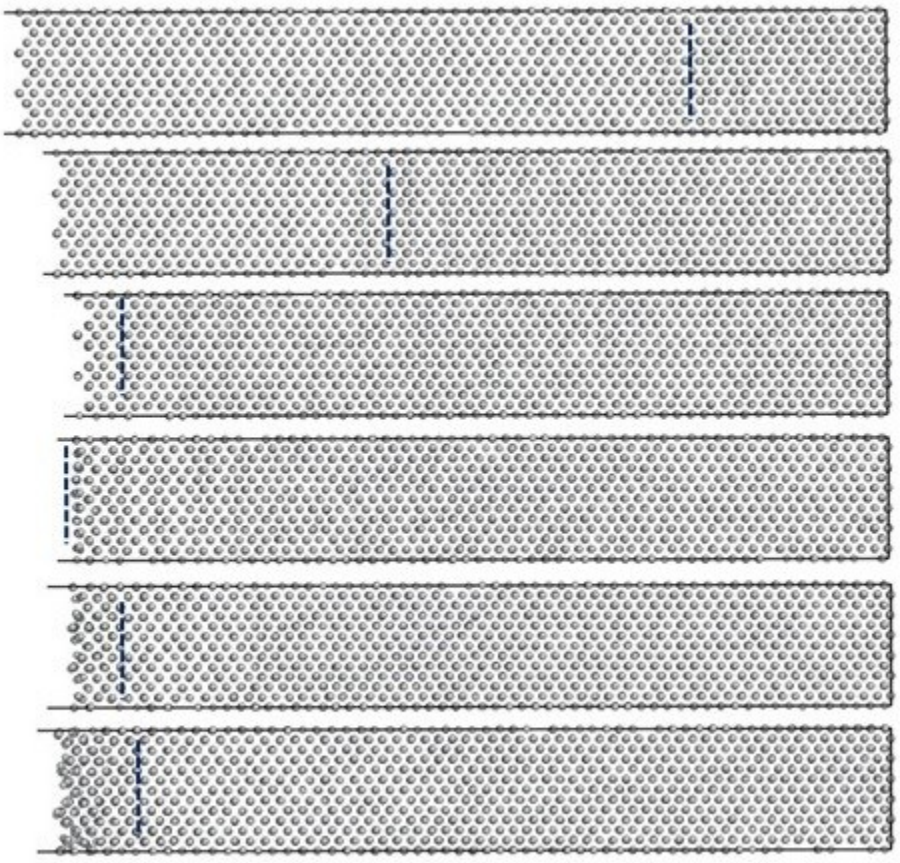

Figure S1. Snapshots from ReaxFF simulations of the collision of a Pt(110) slab against a hard wall positioned at extreme righ-hand-side of the picture. Each snapshot captures the systems at increasing times from top to bottom. The times are 1.2, 2.4, 3.4, 3.9, 4.1, 4.2 psec from top to bottom. Dark blue dotted are added to highlight the position of the shock wave.

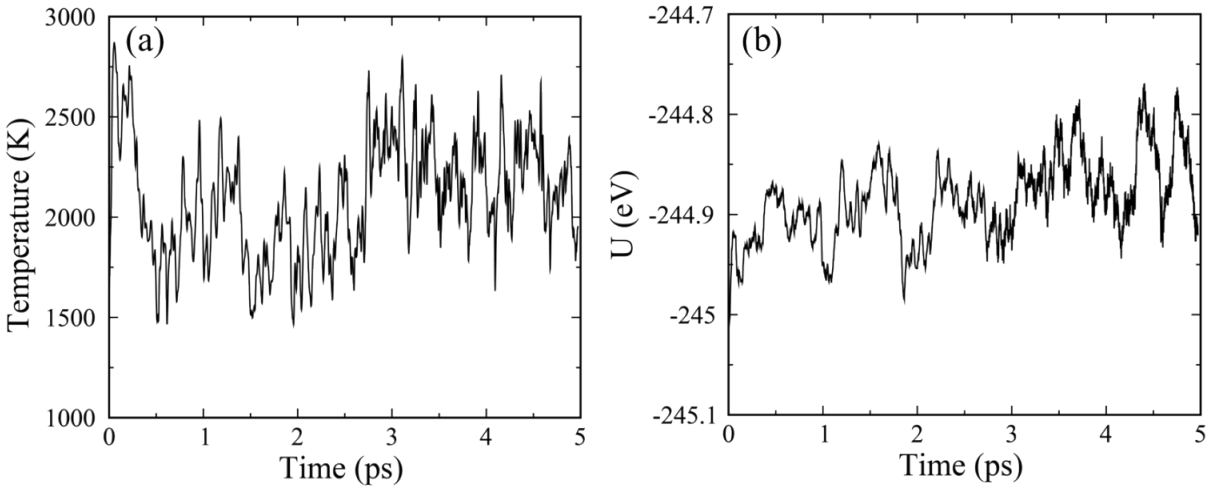

Figure S2. The temperature (a) and internal energy (b) changes during a typical ASW simulation. The initial equilibrium temperature is 1700 K and the average internal energy is -247.91 eV before applying ASW.

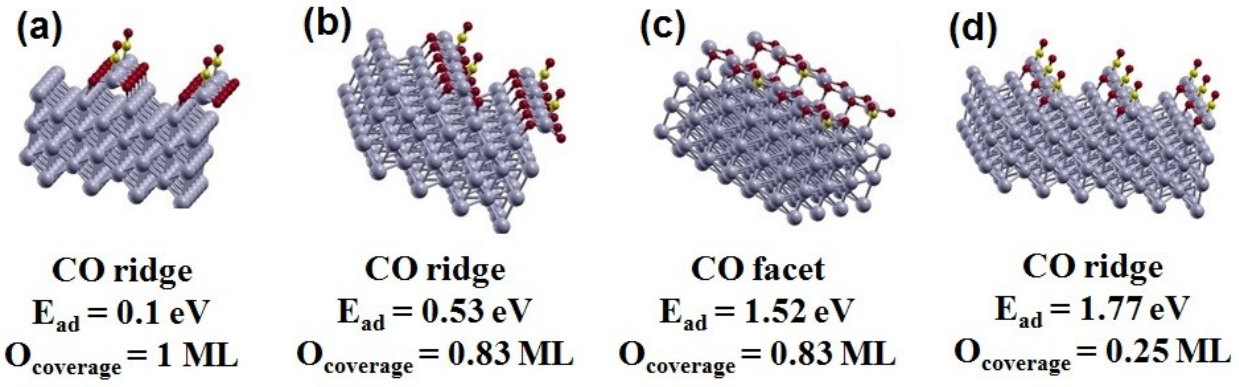

Figure S3. DFT adsorption energies of the CO molecule onto reconstructed Pt(110) surfaces at different coverages illustrated by atomistic pictures as discussed in the main text.

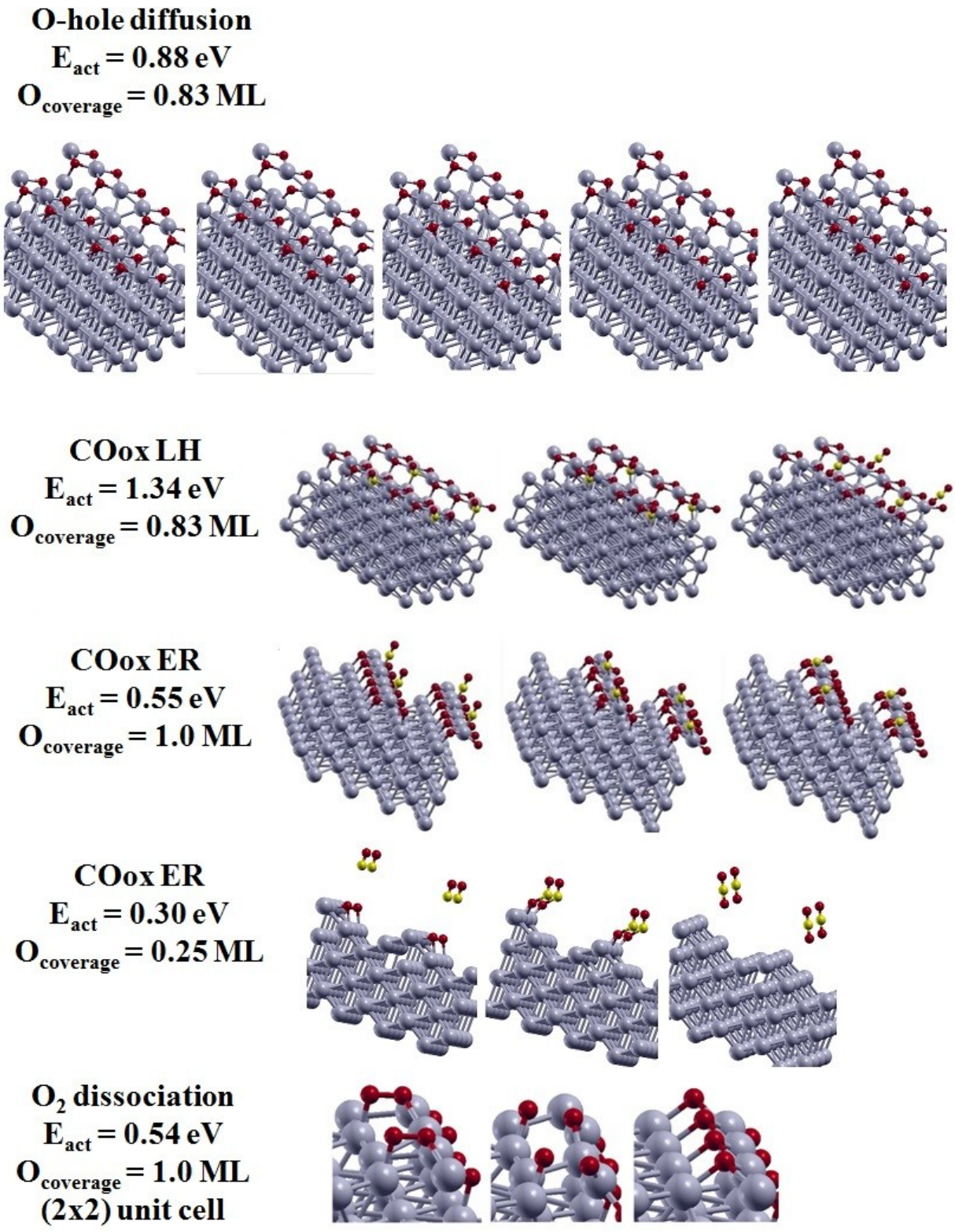

Figure S4. DFT/NEB energy barriers and associated mechanisms illustrated by atomistic pictures onto reconstructed Pt(110) surfaces at different coverages as discussed in the main text.

References

(1) Bengtsson, L. Dipole Correction for Surface Supercell Calculations. *Phys. Rev. B* **1999**, *59*, 12301−12304

(2) Freyer, N.; Kiskinova, M.; Pirug, G.; Bonzel, H. P. Oxygen Adsorption on Pt(110)-(1×2) and Pt(110)-(1×1). *Surf. Sci.* **1986**, *166*, 206−220.

(3) von Oertzen, A.; Mikhailov, A. S.; Rotermund, H. H.; Ertl, G. Subsurface Oxygen in the CO Oxidation Reaction on Pt(110): Experiments and Modeling of Pattern Formation *J. Phys. Chem. B* **1998**, *102*, 4966−4981.